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SPECIAL ISSUE ARTICLE

Size Evolution of Ordered SiGe Islands Grown by Surface Thermal Diffusion on Pit-Patterned Si(100) Surface

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Abstract The ordered growth of self-assembled SiGe islands by surface thermal diffusion in ultra high vacuum from a lithographically etched Ge stripe on pit-patterned Si(100) surface has been experimentally investigated. The total surface coverage of Ge strongly depends on the distance from the source stripe, as quantitatively verified by Scanning Auger Microscopy. The size distribution of the islands as a function of the Ge coverage has been studied by coupling atomic force microscopy scans with Auger spectro-microscopy data. Our observations are consistent with a physical scenario where island positioning is essentially driven by energetic factors, which predominate with respect to the local kinetics of diffusion, and the growth evolution mainly depends on the local density of Ge atoms.

Keywords SiGe islands · Ordering · Pit-patterned Si surface · Nucleation · Diffusion · Growth dynamics

Introduction

Stranski–Krastanov (SK) growth of SiGe islands on Si(100) substrates has become a model system for nucleation [1], faceting [2, 3] and intermixing [4, 5]. For many technological applications [6], which require the individual addressability of the islands, the randomness in their positioning on a flat substrate impose serious limitations. Substrate two-dimensional patterning has been shown to induce an ordered growth process with a controlled positioning [7]. Usually, the templates consist of a 2D array of pits where only a single island develops at the pit bottom [8]. In order to further explore device engineering in these systems, some crucial parameters must be controlled. From a mesoscopic point of view, island shape and size distributions are the most important factors that can be managed. Hence, it is imperative to understand and control the growth conditions for a rational nanostructures design.

In this work, we investigate the self-assembly of SiGe islands on a pit-patterned Si(100) surface grown by a novel method, which makes use of a lithographically etched Ge stripe used as atomic source directly placed on the sample surface. The total surface coverage of Ge strongly depends on the distance from the source stripe, so that the method allows to investigate the island growth over a wide range of dynamical regimes at the same time. A similar approach, using an artificially induced gradient of the Ge coverage, has been recently employed by Brehm et al. [9] for the study of the SK growth onset in case of flat surface. From the size evolution exhibited by the ordered nucleated islands as a function of the distance from the source stripe, we propose a scenario where island positioning is essentially driven by energetic factors, which predominate with respect to the local kinetics of diffusion, and the growth evolution mainly depends on the local density of Ge atoms.

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Experiment and Methods

The samples consist of Ge stripes (width $\sim 3\div 5\text{ }\mu\text{m}$) obtained by a photolithographic patterning of pure Ge thin films (thickness $\sim 50\text{ nm}$), grown on a Si(100) substrate by Low Energy Plasma Enhanced Chemical Vapour Deposition (LEPECVD) [10]. The sample surface close to the stripe region has been patterned with a squared two-dimensional array of circular pits (diameter $\sim 150\text{ nm}$, depth $\sim 25\text{ nm}$, period $\sim 1\text{ }\mu\text{m}$) with an overall width of about $10\text{ }\mu\text{m}$ from the stripe edge, obtained by means of Electron Beam Lithography (EBL) and reactive Ion Etching (RIE). Removal of native silicon oxide and germanium oxide has been obtained by using a diluted HF solution at 10% for 30 s at room temperature. Surface contaminations have been removed by in situ low-temperature outgassing ($T \leq 500^\circ\text{C}$) and a mild Ar^+ ion sputtering. A PHI 660 Scanning Auger Microscope (SAM) has been used for in situ Scanning Electron Microscopy (SEM) and spatially resolved chemical characterization at the sample surface before and after thermal diffusion. The stripes act as Ge sources directly placed on the sample surface, and self-assembled SiGe islands spontaneously originate along a continuous diffusion profile after annealing at high temperatures in UHV. The samples have been annealed by direct Joule heating running a DC current through the Si substrate. The temperature stabilization takes less than 30 s, and the temperature spatial distribution is highly uniform in the investigated area. The base pressure during the annealing time was always better than 1×10^{-9} torr. Atomic Force Microscopy (AFM) for ex situ analysis of the nucleated islands has been performed using a Veeco Innova microscope operated in tapping mode with ultra-sharp tips (nominal tip radius about 2 nm). Statistical

analysis of AFM data has been performed using freely available software tools [11].

Results and Discussion

During the annealing process, the Ge diffusing from the stripe on the Si surface forms a continuous over-layer (OL). Figure 1a shows the SEM micrograph of the stripe in a region of the sample without pit patterning before (upper inset) and after (main panel) a 7.5-min annealing at 625°C . The shading at the sides of the stripe results from the compositional contrast of the secondary electron emission between Ge, diffused on the surface, and Si in the substrate. The thickness and composition of the diffused over-layer, shown in Fig. 1b, have been obtained by monitoring the Ge LMM and Si LMM Auger lines measured as a function of x and fitting their intensities with a discrete layer model [12] where the OL is approximated by a $\text{Si}_{1-x}\text{Ge}_x$ thin film of variable thickness along the x -direction and uniform composition x along its depth [13]. Since the size of the electron beam of our Scanning Auger Microscope is considerably smaller than the distance between the islands and with respect to the length scale over which the thickness and composition vary significantly, we could assume a uniform layer in the probed region, and thus, the Auger spectro-microscopy analysis performed in situ after the annealing allowed the determination of the thickness and composition of the wetting layer between the islands as a function of x .

A gradient into the Ge coverage has been thus induced by the diffusion process, strongly modulating the local density of Ge atoms upon the distance from the source stripe. For annealing at 625°C , we found an average Ge

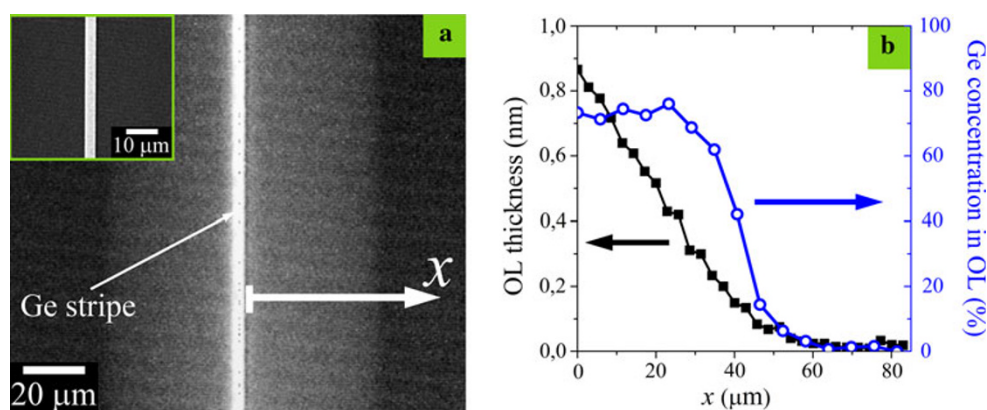


Fig. 1 (Color online). **a** Scanning electron micrograph of the stripe in a region of the sample without pit patterning before (upper inset) and after (main panel) a 7.5-min annealing at 625°C . The lighter part at the center of the image is the Ge stripe. The shading at the sides of the stripe results from the compositional contrast of the secondary electron emission between Ge, diffused on the surface, and Si in the

substrate. **b** Thickness (filled black squares) and composition (open blue circles) of the diffused over-layer upon the distance, x , from the stripe edge, as obtained by monitoring the Ge LMM and Si LMM Auger lines and fitting their intensities with a discrete layer model (see text)

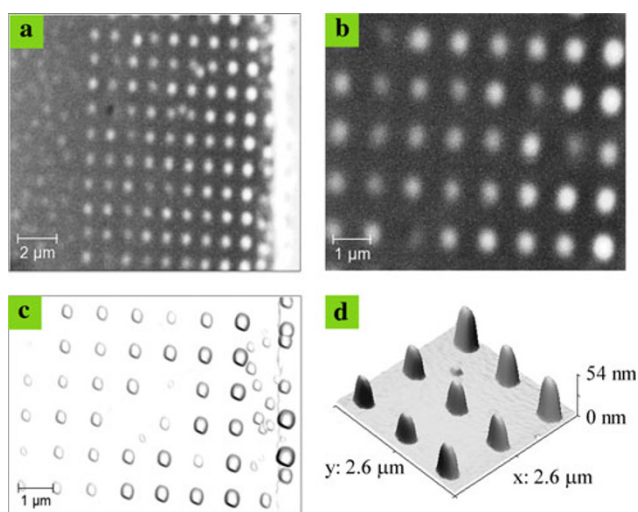


Fig. 2 SEM **a–b** and AFM **c–d** images of the sample surface in the pit-patterned region after annealing at 625°C for 7.5 min. The AFM image in **c** is shown in gradient mode

relative concentration of about 0.73 ± 0.03 , in good agreement with the values found in literature for the case of MBE deposition [14]. Spontaneous nucleation of self-assembled SiGe islands coexists with the continuous surface diffusion of Ge. Figure 2a–b and c–d show representative SEM and AFM images, respectively, of the sample surface in the pit-patterned region after annealing at 625°C for 7.5 min. Islands are essentially dome shaped and preferentially develop at the pit positions creating an ordered squared 2D array following the pit pattern: only $\sim 10\%$ of islands nucleated within the textured region are outside of the pit positions, and only $\sim 7\%$ of pits are empty or partially filled. The ordered island growth has been obtained by controlling the local atomic mobility by purposely choosing the growth parameters (annealing time and temperature). This allowed to make the diffusion pathway run by each atom before the formation of a critical nucleus longer than the step size of the pit pattern and thus to favor the island formation at pit positions, which represent preferential nucleation sites since a total elastic energy minimum is reached at the pit bottom [8].

Figure 3a shows the volume of individual ordered grown islands nucleated in the pit-patterning region, derived by AFM data, as a function of the distance, x , from the stripe. For the estimation of the island volume, we considered only the portion of the dome above the surrounding 2D flat surface; the contribution of Ge volume inside the pit underneath the island is negligible and has not been taken into account. Indeed, our conclusions about the factors governing the growth process will be not affected by this evaluation. Larger islands preferentially nucleate close to the stripe, while small islands grow farther away from it (see Fig. 3a), showing a continuous variation

greater than one order of magnitude in their volume. In this case of ordered growth, the areal density of the nucleated islands and their positioning are essentially driven by the elastic energy minimization on a textured surface, which predominate with respect to the local kinetics of diffusion of Si and Ge atoms. We propose that this size evolution is mainly due to the gradient into the Ge coverage induced by the long-scale diffusing motion of Ge atoms from the stripe. In fact, regions farther away from the stripe exhibit a lower local density of Ge atoms and thus a smaller amount of Ge available for a growing island. A lower average Ge content could be thus responsible for a smaller island size, as experimentally demonstrated by Rastelli et al. [15] in case of randomly nucleated islands grown by Molecular Beam Epitaxy (MBE). However, in case of island growth by Ge surface diffusion over a flat Si(100) surface without any pit patterning, we observed that the region with highest Ge coverage (close to the stripe) presents the lowest average island sizes, while where the coverage decreases to about 4 ML (farther away from the stripe), the biggest average dimensions of the islands are attained (see Fig. 3b, showing the case of annealing at 600°C). Therefore, the artificial pit patterning used in combination with the self-assembled growth by surface thermal diffusion allows to modulate the Ge coverage keeping at the same time fixed the island density and the capture zone area, from where islands gather mass to grow. This effectively separates the factors governing the formation of the critical nuclei from the following growth process of the islands determining their final size. Contrarily, in case of random nucleation, kinetic factors influence both the nucleation mechanism and the growth process, strongly modulating the island density and the capture zone area. Actually, on a flat Si surface, the island density decreases going far away from the stripe (not shown), and thus, the capture zone area increases correspondingly, inducing a progressive increase in the island volume (Fig. 3b), eventually determined by the interplay of Ge coverage, capture area and SiGe intermixing. A more detailed discussion about randomly nucleated islands is far from the scope of the present paper and will be reported elsewhere [13, 16].

A quantitative validation of our educated guess about the factors governing the ordered growth process in presence of pit patterning can be obtained by correlating the volume of ordered grown islands with the effective Ge volume within the OL per island (see Fig. 3c). The last is obtained by integrating the Ge coverage within the capture zone of each island. In principle, in case of a perfectly ordered 2D squared array of islands, the capture zone has the same area for all islands having a squared shape with a side equal to the step size of the pit pattern. However, during data analysis, in order to correctly take into account even the case of not perfectly ordered growth, the capture

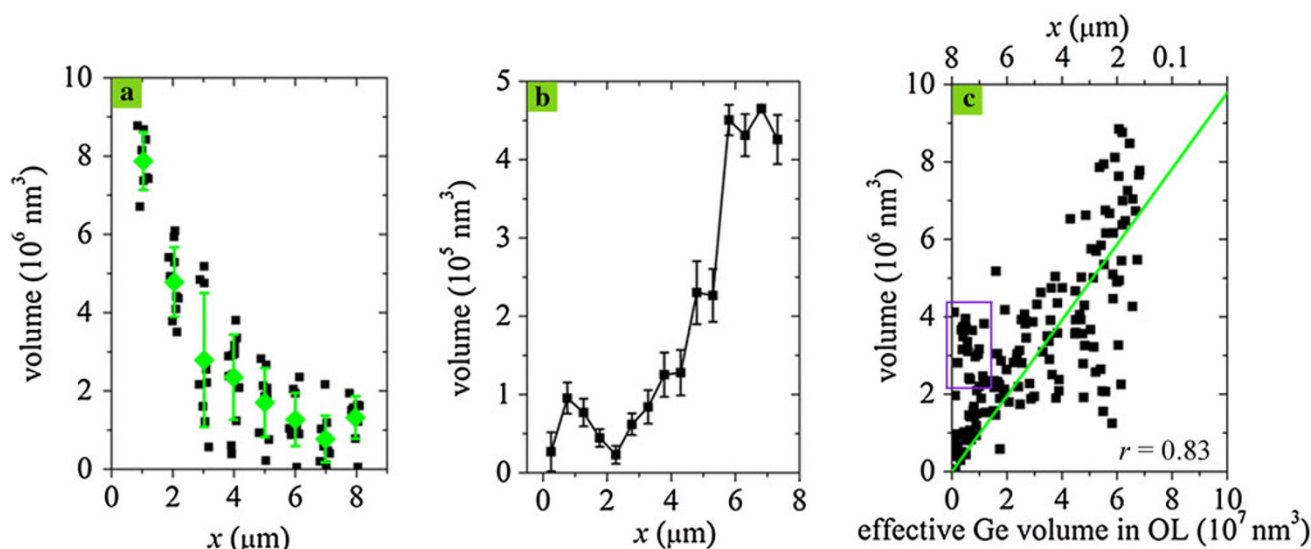


Fig. 3 (Color online). **a** Volume distribution of individual ordered grown islands on a pit-patterned Si(100) surface, derived by AFM scans, as a function of the distance, x , from the stripe. *Green diamonds* represent average values. **b** Volume distribution of randomly nucleated islands on a *flat* Si(100) surface without any pit patterning, as a function of the distance from the stripe in case of annealing at 600°C. **c** Scatter plot of the volume of ordered grown islands as a function of the effective Ge volume within the OL per

island, as obtained by integrating the Ge coverage within the capture zone of each island. The *green curve* is the best linear fitting of the data, and r is the Pearson's coefficient. The *blue square* indicates a set of islands at low Ge coverage, far away from the stripe (see scale on *top* for a coarse indication of the distance), that deviate from the capture zone model, presenting higher volumes with respect to the linear scaling fitting (see text)

zone for each island has been obtained by the Voronoi tessellation of the island network. A good linear correlation has been found between the volume of ordered grown islands and the effective Ge volume within the OL as given by a Pearson's coefficient, r , of about 0.83, confirming that their size evolution as a function of x is mainly driven by the gradient into the Ge coverage. Thus, as a first approximation, the growth process could be described within a capture zone model [17, 18], extended to the case of variable Ge coverage. According to this model, the island volume scales linearly with the integral of the Ge coverage over the capture zone area (instead of the straight capture zone area as in case of homogeneous coverage [18]).

We stress that the above-mentioned model do not exclude SiGe intermixing phenomena during the annealing process, according to which Si penetrates the growing islands. Indeed, the growth process can be still well described by this scaling behavior considering that islands are in a chemical equilibrium with the wetting layer. Furthermore, a detailed observation of Fig. 3c reveals a non-negligible set of islands at low Ge coverage (far away from the stripe) that deviate from the above-mentioned model, presenting higher volumes with respect to the linear scaling fitting. We speculate that the deviation from this linear scaling behavior could be due to an enhanced SiGe intermixing, which become significant in low Ge coverage

regions far away from the stripe. There the timescale of the growth process is slower with respect to the high coverage region close to the stripe, leaving enough time to islands to gather Si from their surroundings. If this hypothesis would be true, we expect that in islands grown farther away from the stripe, the alloying offers a path for a partial elastic strain relaxation increasing the critical size for insertion of misfit dislocations, as found in the case of random nucleation [19]. Opposite to this, the almost negligible SiGe intermixing for islands grown close to the stripe due to a rapid growth process should be accompanied by a plastic relaxation by misfit dislocations. The experimental verification of these hypothesis overcomes the scope of the present paper, but could motivate a further investigation into the evolution of Ge composition and strain state of islands grown by surface thermal diffusion on a pit-patterned surface, possibly addressed in future by Micro-Raman Spectroscopy and Transmission Electron Microscopy.

Conclusions

In conclusion, we have investigated the growth of self-assembled SiGe islands on a pit-patterned Si(100) surface by surface thermal diffusion from a lithographically etched Ge stripe. The ordered island growth has been obtained by

controlling the local atomic mobility and the length of the diffusion pathway of Ge atoms, by means of a correct choice of the growth parameters (annealing time and temperature). A controlled size evolution of islands grown in a ordered squared 2D array has been obtained by controlling the diffusion dynamics of Ge from the source stripe. This entails a variation of the local density of Ge atoms upon the distance from the source able to modulate the average Ge content inside the growing islands and thus their dimensions.

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